A MULTISCALE MODEL FOR THE EFFECTIVE THERMAL CONDUCTIVITY TENSOR OF A STRATIFIED COMPOSITE MATERIAL[1]

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ABSTRACT

The thermal modelling of composites has three essential objectives : (i) the comprehension of their thermal behaviour ; (ii) the composite scaling in order to satisfy specific requirements ; (iii) the optimal analysis of experimental results from thermal characterisation. For a complete study of the material, each of these three points must be taken into account at the fibre scale (≈ 10 μm), the yarn scale (≈ 1 mm), and the composite scale (≈ 10 cm). This work presents a multi-scale modelling of the effective thermal conductivity tensor of a stratified composite material. The longitudinal and transverse thermal conductivities of the yarn are computed from optical microscopic imaging of the material. The isotropic thermal conductivity of the loaded matrix is computed by the Bruggeman model. Then, the thermal conductivity tensor is performed by a finite element method taking into account the morphology of the fabric. Computed values are close to experimental values measured by classical methods. Finally, analytical relations are proposed to obtain an efficient model which can be used in a multi-phenomena simulation of the composite structure.

1. INTRODUCTION

Woven-fabric/polymer matrix composites have been extensively studied because of the relative ease and the low cost of their manufacturing. Many industrial issues are due to their very good specific mechanical properties. However, for the thermal protection applications, charring ablators are the most widely used. Are generally used phenolic, epoxy or silicon resins with glass or carbon fibres. In this case, the thermal properties of the composites is very important.

The purpose of this paper is to describe one example of modelling of the effective density, effective specific heat and effective thermal conductivity tensor of a carbon woven-fabric/phenolic matrix composite. The strategy is to take into account the measured properties of each constituent (fibres, matrix, fillers) and the woven-fabric of the composite. Both analytical and numerical approaches are described.

The measured data used in the different models and the computation results are presented. Then the difference between these results and the experimental data on the composites is discussed.

2. EXPERIMENTAL DATA FOR THE COMPOSITE AND ITS CONSTITUENTS

2.1. Morphological characteristics of the composite material

The material under consideration is made from based rayon carbon fibres (Fig. 1, medium diameter: d=12 μm ; volumetric fraction: $\alpha_{f,C}$=0.42) and phenolic resin matrix. Carbon loads (volumetric fraction: $\alpha_{l,C}$=0.06) are imbedded into the inter-yarn matrix, but not into the intra-yarn matrix. The porosity (ε=0.02) is uniformly distributed into the material. The yarns, identical for chain and weft directions (720 fibres per yarn, volumetric fraction of fibres into the yarn : $\alpha_{f,Y}$=0.6), are woven in order to make plies

of satin 8/3 (Fig. 2). These plies are then put on each other, without disorientation, to constitute the stratified composite.

2.2. Thermophysical properties of the constituents

The density of the fibres, the resin, and the loads: ρ_f=1800 kg.m^{-3}, ρ_r=1300 kg.m^{-3} and ρ_l=2200 kg.m^{-3}, are measured by Helium pycnometry (commercial apparatus : Accupyc 1330, Micromeritics) with an uncertainty lower than 3 %. The thermal conductivities (longitudinal: $\lambda_{f,L}$=6 W.m^{-1}.K^{-1}, and transverse: $\lambda_{f,T}$=1.6 W.m^{-1}.K^{-1}) of the fibres are obtained from thermal diffusivity measurements, at room temperature, by a photothermal microscopy method [1]. These very difficult measurements are performed with an estimated uncertainty of 20 %. The thermal conductivity of the resin, λ_r=0.4 W.m^{-1}.K^{-1}, is also determined from diffusivity measurements realised on a bulk sample by the flash method [2]. The thermal conductivity of the loads, λ_l=100 W.m^{-1}.K^{-1}, is equally determined from diffusivity measurements on loaded matrix samples at various concentrations. These measurements are performed with a large uncertainty of 20 %. Finally, the specific heat of carbon fibres, phenolic resin and carbon loads are obtained by differential scanning calorimetry (commercial apparatus : DSC, Setaram) with an estimated uncertainty of 10 % : c_f=750 J.kg^{-1}.K^{-1}, c_l=1050 J.kg^{-1}.K^{-1}, c_r=600 J.kg^{-1}.K^{-1}.

2.3. Thermophysical properties of the composite material

The thermophysical properties of the composite are determined with identical techniques. The density, ρ_C=1490 kg.m^{-3} is measured with an uncertainty of 3 %. The thermal conductivities, $\lambda_{C,//}$=1.81 W.m^{-1}.K^{-1} in the parallel direction and $\lambda_{C,\perp}$=1.16 W.m^{-1}.K^{-1} in the perpendicular direction, are determined from diffusivity

measurements with an uncertainty of 10%. Finally, the specific heat, c_C=900 J.kg^{-1}.K^{-1} is measured by DSC with an uncertainty of 10%.

3. ANALYTICAL MODEL FOR THE THERMOPHYSICAL EFFECTIVE PROPERTIES

3.1. Volumetric fractions

The volumetric fraction of the fibres into the yarns, $\alpha_{f,Y}$=0.6, is obtained by analysis of yarn cross-section photographs. Consequently, the volumetric fraction of the resin into the yarns is simply determined by: $\alpha_{r,Y} = 1 - \alpha_{f,Y} = 0.40$. The volumetric fraction of the loads into the matrix is also determined using algebrical relations between the volumetric fractions of all the constituents :

$$\alpha_{l,M} = \frac{\alpha_{l,C}}{1 - \dfrac{\alpha_{f,C}}{\alpha_{f,Y}} - \varepsilon}$$

or numerically: $\alpha_{l,M} = 0.21$. Consequently, the volumetric fraction of the resin into the loaded matrix is simply determined by: $\alpha_{r,M} = 1 - \alpha_{l,M} = 0.79$, the volumetric fraction of the resin into the composite by: $\alpha_{r,C} = 1 - \alpha_{f,C} - \alpha_{l,C} - \varepsilon = 0.50$, the volumetric fraction of the yarns into the composite by: $\alpha_{Y,C} = \alpha_{f,C}/\alpha_{f,Y} = 0.70$, and the volumetric fraction of the loaded matrix into the composite by: $\alpha_{M,C} = 1 - \alpha_{Y,C} - \varepsilon = 0.28$.

3.2. Effective density of the composite

The effective density of a material constituted by N constituents of volumetric fraction α_i is given by: $\rho = \sum_{i=1}^{N} \alpha_i \rho_i$. Using this definition to determine the effective density of the yarns and the loaded matrix, the effective density of the composite is given by:

$$\rho_C = \alpha_{Y,C}(\alpha_{f,Y}\rho_f + \alpha_{r,Y}\rho_r) + \alpha_{M,C}(\alpha_{l,M}\rho_l + \alpha_{r,M}\rho_r)$$

or numerically: $\rho_C = 1538$ kg.m^{-3}.

3.3. Effective specific heat of the composite

The effective specific heat of a material constituted by N constituents of volumetric

fraction α_i is given by: $c = \sum_{i=1}^{N} \alpha_i \rho_i c_i \bigg/ \sum_{i=1}^{N} \alpha_i \rho_i$. Using this definition to determine the

effective specific heat of the yarns and the loaded matrix, the effective specific heat of

the composite is given by :

$$c_C = \frac{\alpha_{Y,C}(\alpha_{f,Y}\rho_f c_f + \alpha_{r,Y}\rho_r c_r) + \alpha_{M,C}(\alpha_{l,M}\rho_l c_l + \alpha_{r,M}\rho_r c_r)}{\alpha_{Y,C}(\alpha_{f,Y}\rho_f + \alpha_{r,Y}\rho_r) + \alpha_{M,C}(\alpha_{l,M}\rho_l + \alpha_{r,M}\rho_r)}$$

or numerically: $c_C = 864$ J.kg^{-1}.K^{-1}.

4. NUMERICAL MODEL FOR THE EFFECTIVE THERMAL CONDUCTIVTY TENSOR

4.1. Effective longitudinal and transverse conductivities of the yarns

The fibres being aligned in the yarns, their effective longitudinal conductivity is simply

determined by a parallel model :

$$\lambda_{Y,L} = \alpha_{f,Y}\lambda_{f,L} + \alpha_{r,Y}\lambda_r$$

or numerically: $\lambda_{Y,L} = 3.76$ W.m^{-1}.K^{-1}. The effective transverse conductivity can be

determined by different methods. A frame of this property is given by the Hashin-

Striktman model [3]: $0.85 < \lambda_{Y,T} < 0.98$ W.m^{-1}.K^{-1}, whereas Rayleigh [4] and

Bruggeman [5] models can also be used to provide approximate values, respectively:

$\lambda_{Y,T} = 0.85$ W.m^{-1}.K^{-1} and $\lambda_{Y,T} = 0.89$ W.m^{-1}.K^{-1}. Moreover, the cross-section

photographs used to determine the volumetric fraction of the fibres into the yarns can be

used to compute their effective transverse conductivity by a direct method. In this

method, a « hot » temperature $T_H=1$ is imposed on one boundary of the medium, a « cold » temperature $T_C=0$ is imposed on the opposite boundary, and an isolation condition is imposed on both other boundaries (Fig. 3), in order to compute the temperature field in the material and to deduce the effective conductivity by the relation:

$$\lambda = \frac{\Phi\, e}{T_H - T_C}$$

where Φ represents the heat flow and e the distance between imposed temperature boundaries. Using this method, the transverse conductivity of the yarn can be evaluated : $\lambda_{Y,T} = 0.89$ W.m^{-1}.K^{-1}. Finally, the direct method can also be applied on regular arrays of cylinders (Fig. 4). These computations lead for square array to: $\lambda_{Y,T} = 0.86$ W.m^{-1}.K^{-1} and for hexagonal array to: $\lambda_{Y,T} = 0.85$ W.m^{-1}.K^{-1}. All the values computed for the effective transverse conductivity of the yarns are very close. For its simplicity, the Bruggeman model has been chosen to calculate $\lambda_{Y,T}$:

$$\lambda_{Y,T} = \lambda_r \frac{\left(1-\alpha_{f,Y}\right)^2 \left(\frac{\lambda_{f,T}}{\lambda_r}-1\right)^2 + 2\frac{\lambda_{f,T}}{\lambda_r} - \sqrt{\left[\left(1-\alpha_{f,Y}\right)^2 \left(\frac{\lambda_{f,T}}{\lambda_r}-1\right)^2 + 2\frac{\lambda_{f,T}}{\lambda_r}\right]^2 - 4\left(\frac{\lambda_{f,T}}{\lambda_r}\right)^2}}{2}$$

This relation leads to : $\lambda_{Y,T} = 0.89$ W.m^{-1}.K^{-1}.

4.2. Effective conductivity of the loaded matrix

The volumetric fraction of loads being relatively low ($\alpha_{l,M} = 0.21$), the effective conductivity of the loaded matrix can be determined by the Maxwell-Eucken model [7] with a good approximation :

$$\lambda_M = \lambda_r \frac{1 + 2\alpha_{l,M} \dfrac{1 - \dfrac{\lambda_r}{\lambda_{ch}}}{1 + 2\dfrac{\lambda_r}{\lambda_{ch}}}}{1 - \alpha_{l,M} \dfrac{1 - \dfrac{\lambda_r}{\lambda_{ch}}}{1 + 2\dfrac{\lambda_r}{\lambda_{ch}}}}$$

This relation leads to : $\lambda_M = 0.72$ W.m^{-1}.K^{-1}.

4.3. Effective thermal conductivity tensor of the composite

The calculation of the effective thermal conductivity of the composite is realised in two

steps. At first, the material is supposed to be non porous. So, the volumetric fraction of

yarn in the composite becomes: $\overline{\alpha}_{Y,C} = \alpha_{Y,C}/(1-\varepsilon) = 0.71$. The conductivity tensor is

determined by the direct method applied on a periodic pattern of the ply (Fig. 5). The

morphology (yarn cross-section and yarn spacing) of the latest is determined in order to

represent the real material and to correspond with the volumetric fraction of the yarns

into the composite. The thermal problem is solved by the finite element method

(Software CAST3M, CEA, France). The temperature difference T_H-T_C is successively

applied along the three directions. From these three numerical experiments, the effective

conductivity tensor of the composite is computed by the relation [8]:

$$\lambda_{i,j} = \frac{\langle \Phi_i \rangle}{\langle \partial T/\partial x_j \rangle}$$

where $\langle \Phi_i \rangle$ represents the mean heat flow parallel to the x_i direction, and $\langle \partial T/\partial x_j \rangle$ the

mean temperature gradient in the x_j direction.

Finally, the material is supposed to be porous. The porosity being low ($\varepsilon = 0.02$), the

Maxwell model [9] provides a good approximation of the effective composite

conductivities:

$$\lambda_{C,//} = \overline{\lambda}_{C,//} \, 2 \frac{1-\varepsilon}{2+\varepsilon} \qquad \lambda_{C,\perp} = \overline{\lambda}_{C,\perp} \, 2 \frac{1-\varepsilon}{2+\varepsilon}$$

These relations lead to : $\lambda_{C,//} = 1.78 \, \text{W.m}^{-1}.\text{K}^{-1}$ and $\lambda_{C,//} = 0.81 \, \text{W.m}^{-1}.\text{K}^{-1}$.

5. ANALYTICAL MODEL FOR THE EFFECTIVE THERMAL CONDUCTIVITY TENSOR

The model developed at the previous paragraph uses analytical relations to compute the effective conductivities of the yarns and the loaded matrix, and numerical relations to compute the effective conductivity tensor of the composite. The objective of this paragraph is to propose approximate analytical models to compute the effective conductivity tensor of the composite.

5.1. Effective conductivity in the direction parallel to ply

At first, the material is supposed to be non porous. The ply is decomposed into two parts: one containing the yarns parallel to the chain direction, and the other containing the yarns parallel to the weft direction. The thermal conductivity of the first part, in the direction parallel to the chain yarns, is called λ_1, the one of the second part is called λ_2. The conductivity λ_1 can be evaluated by a parallel model:

$$\lambda_1 = \overline{\alpha}_{Y,C} \, \lambda_{Y,L} + (1-\overline{\alpha}_{Y,C}) \, \lambda_M$$

whereas the conductivity λ_2 can be evaluated by a serial model:

$$\lambda_2 = \frac{\lambda_r \, \lambda_{Y,T}}{\overline{\alpha}_{Y,C} \, \lambda_M + (1-\overline{\alpha}_{Y,C}) \, \lambda_{F,T}}$$

The association of both parts in a parallel scheme provides the effective conductivity of the non porous composite material:

$$\overline{\lambda}_{C,//} = \frac{1}{2} \left[\overline{\alpha}_{Y,C} \, \lambda_{Y,L} + (1-\overline{\alpha}_{Y,C}) \, \lambda_M + \frac{\lambda_M \, \lambda_{Y,T}}{\overline{\alpha}_{Y,C} \, \lambda_M + (1-\overline{\alpha}_{Y,C}) \, \lambda_{Y,T}} \right]$$

Then, using the Maxwell model [9], the effective conductivity of the porous composite material is given by:

$$\lambda_{C,//} = \frac{1-\varepsilon}{2+\varepsilon}\left[\overline{\alpha}_{Y,C}\,\lambda_{F,L} + (1-\overline{\alpha}_{Y,C})\,\lambda_M + \frac{\lambda_M\,\lambda_{Y,T}}{\overline{\alpha}_{Y,C}\,\lambda_M + (1-\overline{\alpha}_{Y,C})\,\lambda_{Y,T}}\right]$$

These relations lead to : $\lambda_{C,//} = 1.82\ \mathrm{W.m^{-1}.K^{-1}}$

5.2. Effective conductivity in the direction perpendicular to ply

At the beginning, the material is again supposed to be non porous. The ply is decomposed into two parts: one containing the yarns parallel to the chain direction, and the other containing the yarns parallel to the weft direction. Considered in the direction perpendicular to the ply, both parts have the same conductivity: $\overline{\lambda}_{C,\perp}$, which can be evaluated by a parallel model:

$$\overline{\lambda}_{C,\perp} = \overline{\alpha}_{Y,C}\,\lambda_{Y,T} + (1-\overline{\alpha}_{Y,C})\,\lambda_M$$

Then, using the Maxwell model [9], the effective conductivity of the porous composite material is given by:

$$\lambda_{C,//} = \frac{1-\varepsilon}{2+\varepsilon}\left[\overline{\alpha}_{Y,C}\,\lambda_{Y,T} + (1-\overline{\alpha}_{Y,C})\,\lambda_M\right]$$

These relations lead to : $\lambda_{C,//} = 0.82\ \mathrm{W.m^{-1}.K^{-1}}$.

6. DISCUSSION

The data of the problem (morphological characteristics and properties of the composite materials) are indicated with associated uncertainties in the first paragraph. These uncertainties on the data lead to uncertainties on the results. For each calculated property, both lower and upper values are computed in order to determine mean value and uncertainty range (Tab. I). The bounds of the range are calculated for a

minimisation or a maximisation of the property value due to data uncertainties. For example, a lower value of the composite density is obtain for lower values of constituent densities and volumetric fractions, and higher value of porosity. As a consequence, the mean value of the uncertainty range may be a bit different from the nominal value calculated in the previous paragraph.

The mean value calculated for the density is a good estimation of the measured value. Nevertheless, due to the relative errors accumulation, the uncertainty attached to this result is twice the measured one. The mean value calculated for the specific heat is also a good estimation of the measured value, with a similar uncertainty. For the effective conductivity parallel to ply, the three mean values are similar; the analytical model value being very close to the measured value. Once again, the uncertainties attached to the numerical results are twice the measured one.

Finally, for the effective conductivity perpendicular to ply, both mean numerical results are close together but are also lower than the measured value. This difference indicates that the modelled material is less conductor than the real material. In both analytical and numerical models, as in the real material, the effective longitudinal conductivity of the yarn has a very little influence on the effective conductivity perpendicular to the ply: the periodic pattern indicates a few intertwine between chain and weft yarns. This behaviour is also observed in the mechanical studies of composite materials [10]. So, the effective conductivity perpendicular to the ply mainly depends on the effective transverse conductivity of the yarns, on the effective conductivity of the loaded matrix and on the thermal contact between these elements. In both models, the thermal contact between yarns and loaded matrix is supposed to be perfect. So, it cannot limit the heat transmission across the material. The relatively few volumetric fraction of the loads in

the matrix involves an increase of the resin thermal conductivity but the effective conductivity obtained ($\lambda_M=0.72$ W.m^{-1}.K^{-1}) may not exceed 0.8-0.9 W.m^{-1}.K^{-1}. As a consequence, the difference observed between calculated and measured values of $\lambda_{C,\perp}$ is mainly attributed to a lower transverse effective conductivity of yarns ($\lambda_{Y,T}=0.89$ W.m^{-1}.K^{-1}). This conductivity is calculated with six different models (paragraph 4.1) which provide similar numerical values (from 0.85 to 0.89 W.m^{-1}.K^{-1}). The thermal conductivity of the resin and the volumetric fraction of fibres into the yarns being better known than the thermal properties of the fibre, it may be concluded that the transverse conductivity of the fibre is probably lower than its true value.

7. CONCLUSION

Both analytical and numerical models have been developed in order to compute effective density, effective specific heat and effective thermal conductivity tensor of a composite material. These models were used to calculate the thermophysical properties of a satin 8/3 stratified composite. A good agreement was obtained for all the predicted properties except the effective conductivity in the perpendicular to ply direction. A discussion has then showed that the difference observed between calculated and measured values may be mainly attributed to a lower transverse conductivity of the yarns or, consequently, of the fibres. This problem will be studied by the thermal characterisation of impregnated yarns, in a future work.

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Table I. Numerical results with absolute uncertainties

Effective property of the composite material	Measurement	Numerical model	Analytical model
density, ρ_C (kg.m^{-3})	1490 ± 45	-	1510 ± 89
specific heat, c_C (J.kg^{-1}.K^{-1})	900 ± 90	-	872 ± 105
conductivity parallel to ply, $\lambda_{C,//}$ (W.m^{-1}.K^{-1})	1.81 ± 0.18	1.78 ± 0.38	1.82 ± 0.39
conductivity perpendicular to ply, $\lambda_{C,\perp}$ (W.m^{-1}.K^{-1})	1.16 ± 0.12	0.81 ± 0.13	0.81 ± 0.13

Figure Captions

Fig. 1. Rayon-based carbon fibre.

Fig. 2. Composite material under consideration in this work. Chain yarns cross-sections and weft yarns in horizontal direction.

Fig. 3. Determination of the effective transverse conductivity of the yarns by direct method using yarn cross-section photographs.

Fig. 4. Determination of the effective transverse conductivity of the yarns by direct method using regular arrays of cylinders.

Fig. 5. Determination of the effective conductivity tensor of the composite by direct method applied on one periodic pattern of the ply.
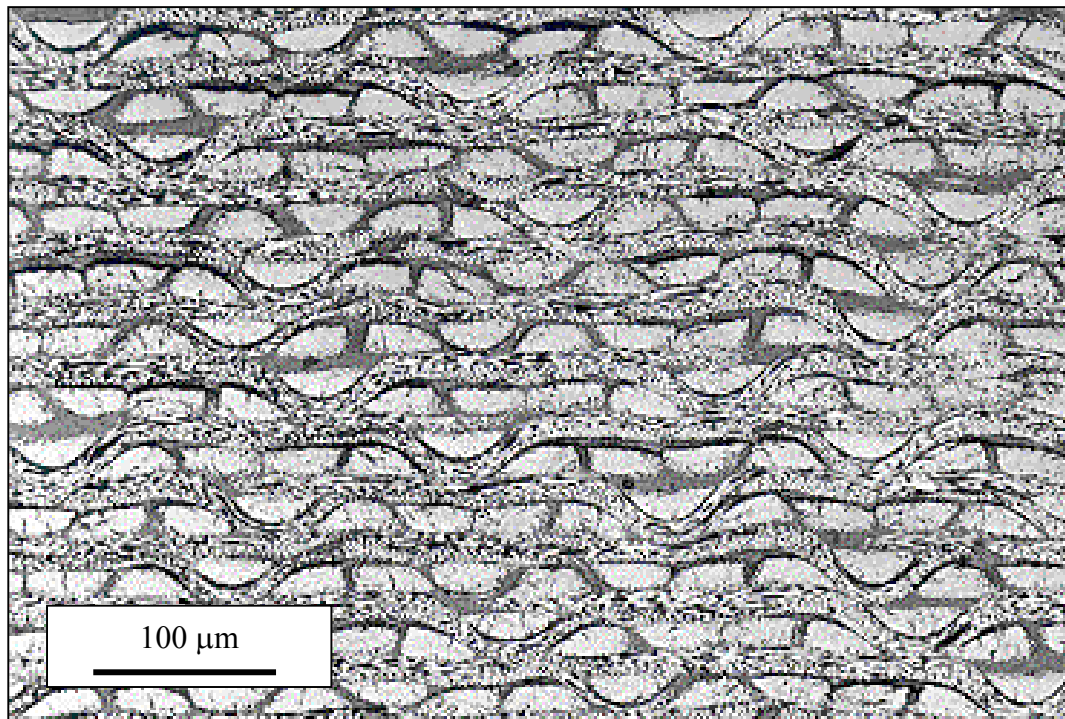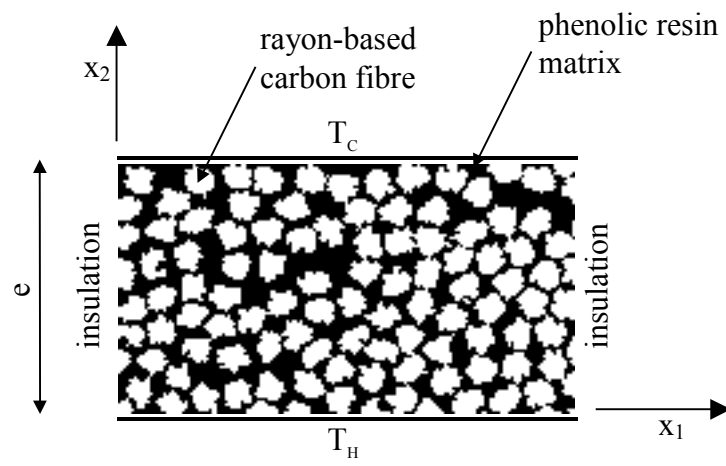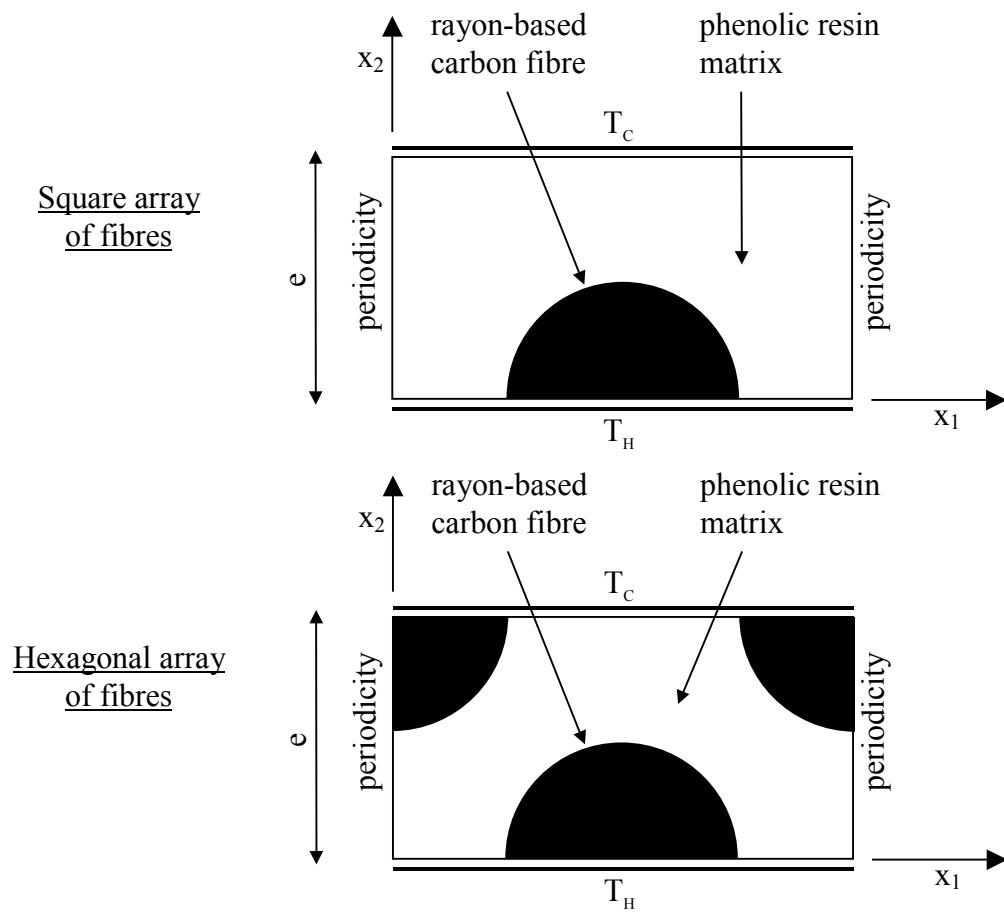
10 µm

Fig. 1

100 μm

Fig. 2

Fig. 3

Square array of fibres

Hexagonal array of fibres

Fig. 4

weft yarns

x_3

periodicity

x_2

T_C

chain yarns

T_H

periodicity

x_1

Fig. 5